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ScienceDirect
Physics Procedia 3 (2010) 593–603
Physics Procedia
www.elsevier.com/locate/procedia

International Congress on Ultrasonics, Universidad de Santiago de Chile, January 2009

Ultrasonic material characterization using diffraction-free PVDF receivers

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Abstract

This work describes the use of a large aperture PVDF receiver in the measurement of density of liquids and elastic constants of composite materials. The density measurement of several liquids is obtained with the accuracy of less than 0.2% using a conventional NDT emitter transducer and a 70-mm diameter, 52-μm P(VDF-TrFE) membrane with gold electrodes. The determination of the elastic constants of composite materials is based in the measurement of phase velocity. It is shown that the diffraction can lead to errors around 1% in the velocity measurement when using a pair of ultrasonic transducers (1MHz and 19mm diameter) operating in transmission-reception mode separated by a distance of 100 mm. This effect is negligible when using a pair of 10-MHz transducers. On the other hand, the dispersion at 10 MHz can result in errors of about 0.5%, measuring the velocity in composite materials. The use of an 80-mm diameter, 52-μm thick PVDF membrane receiver allows measuring the phase velocity without the diffraction effects.

keywords: density, elastic constant, diffraction-free receiver, PVDF, material characterization

1. Introduction

The determination of material properties by ultrasound has been studied by several researchers using the measurement of acoustic parameters, such as: the acoustic impedance, the propagation velocity and the attenuation coefficient. The accuracy in the measurement of these properties is highly dependent on the precision of the phase and the amplitude measurements of the propagating wave. One of the major factors that introduce errors, when measuring the reflection and transmission coefficients, and velocity and attenuation, is the acoustic diffraction. Mathematical correction of the diffraction effect has been successfully applied when using piston-like transducers and calculating its effective radiating aperture [1].

A good physical understanding of the acoustical field pattern is provided by considering the plane and edge waves radiated by a circular transducer. A circular source radiates a plane wave into the geometrical region straight

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doi:10.1016/j.phpro.2010.01.076

ahead of the source, and a spreading edge wave from the periphery of the source. It is clear that diffraction effects can be considered to be caused by the edge wave component of the field. This effect increases as the frequency decreases for the same transducer diameter. In practical transducers, discrepancies from piston-like behavior can be observed, such as a head-wave [2], which originates from a plate wave propagating laterally across the face of the transducer. There are further complications when the field is produced by focusing transducers.

It can be shown that an infinite-plane receiver with uniform sensitivity yields a plane-wave-only measurement [3]. It acts as a spatial filter, responding only to those field components that are perpendicularly incident. In practice this receiver is obtained by using a piezoelectric PVDF (Polyvinylidene Fluoride) or its copolymer (P(VDF-TrFE)) thin-film receiver, sufficiently large to intercept the entire propagating wave and electroded throughout its entire extent [4]. This technique avoids the beam diffraction effect that occurs when using limited size ultrasonic transducers. This is also valid for fields generated by focused transducers, given a distance-independent output in a very low attenuating medium.

This work describes the use of a large aperture PVDF receiver for measuring the density of liquids and the elastic constants of composite materials. The ultrasonic measurement of liquid density based on the reflection coefficient has been performed using different arrangements [5], [6], [7], [8]. This work shows the density measurement of several liquids obtained with accuracy better than 0.2% with a measurement cell using a conventional NDT transducer as an emitter and a 70-mm diameter, 52-μm P(VDF-TrFE) membrane as a receiver [9].

The determination of elastic constants of anisotropic materials by measurement of the density and ultrasonic velocities has been studied by several researchers in the last four decades [10], [11], [12]. The accuracy in the determination of the elastic constants is highly dependent on the precision of the velocity measurement. The effects of diffraction and velocity dispersion are experimentally analyzed using a large aperture PVDF receiver together with a piezoelectric ceramic emitter in through-transmission method of ultrasonic velocity measurement in solid material plates immersed in water. The immersion through-transmission method is based on the measurement of a time delay between the time spent by the wave traveling in the absence of the sample material and the time with the sample material. This time delay is used to calculate the phase velocity and the refraction angle in the sample [11].

The elastic constants of a unidirectional CFRP (carbon fiber reinforced polymer) plate are determined using phase velocities measured with an angle beam through-transmission assembly using the diffraction-free PVDF receiver.

2. Applications of diffraction-free PVDF receivers

2.1. Density

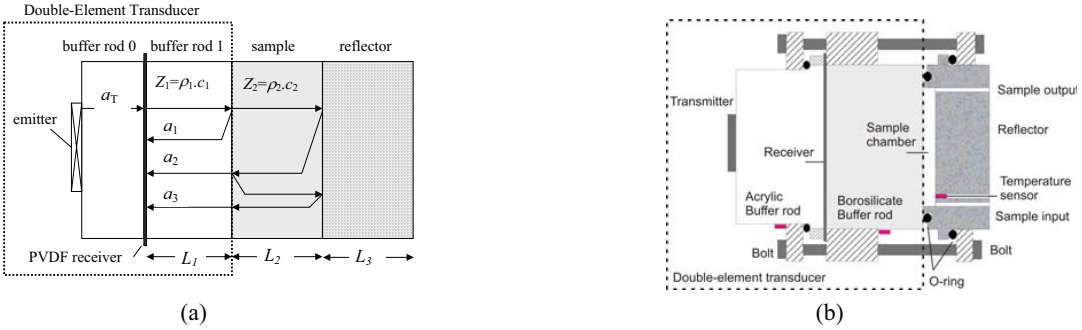
The density ρ of a Newtonian liquid is obtained from the measurement of the characteristic acoustic impedance Z and the propagation velocity c through the relation $Z = \rho c$. The acoustic impedance Z is calculated from the measurement of the reflection coefficient at an interface between a reference material and the liquid.

2.1.1. Density measurement cell

The density measurement cell is depicted in Fig. 1 and 2. It is physically mounted in two parts, in what is called Double-Element Transducer (DET) and sample chamber/reflector. The DET has a ceramic or composite emitter and a large-aperture P(VDF-TrFE) receiver with gold electrodes, separated by an acrylic buffer rod, of length L_0. An additional glass buffer rod (BF1, length L_1) is placed between the receiver and the sample. The stainless-steel reflector (length L_3) incorporates the sample chamber (length L_2) and is bolted to the DET. Platinum temperature sensors are placed in the sample chamber and on the buffer rods.

The measurement method is called the multiple reflections method: the emitter is excited with a short pulse, which propagates through the acrylic buffer rod and is detected by the receiver as $a_T(t)$. This pulse reaches the BF1/sample interface, is partially reflected and reaches the receiver as $a_1(t)$. Subsequent multiple reflections at the sample/reflector interface are detected as signals $a_2(t)$ and $a_3(t)$. The measurement cell dimensions (lengths and

diameters) are chosen in order to avoid the superposition of spurious reflections on the desired signals. For the measurement cell depicted in Fig. 2, the diameter is 138 mm, L_0= 13.5 mm, L_1= 37 mm, L_2= 2 mm and L_3= 30 mm.



(a) (b)

Fig.1 Ultrasonic density measurement cell: a) Schematic representation, b) Mechanical construction.

(a) (b)

Fig.2 Density measurement cell: DET and sample chamber/reflector.

The reflection coefficient at the glass buffer rod/sample interface is:

$$R_{12} = (Z_2 - Z_1)/(Z_2 + Z_1) \tag{1}$$

where $Z_i=\rho_i c_i$, i=1,2, are the specific acoustic impedances of the glass buffer rod and liquid sample, respectively, ρ is density and c is propagation velocity. The sample density ρ_2 is calculated from:

$$\rho_2 = \rho_1 c_1 (1 + R_{12}) / c_2 (1 - R_{12}) \tag{2}$$

The BF1 density ρ_1 is known, and the propagation velocities are calculated from $c_i=2L_i/\Delta t_i$, where Δt_1 is the time delay between $a_T(t)$ and $a_1(t)$, and Δt_2 is the time delay between $a_1(t)$ and $a_2(t)$. These time delays are calculated by using the Hilbert transform of the autocorrelation function [9]. The glass buffer rod length L_1 is known and L_2 is the sample chamber length that is measured from a calibration with distilled water.

The reflection coefficient is obtained from [5]:

$$R_{12} = (1 + x)^{-1/2} \tag{3}$$

where x is calculated by [9]:

$$x = \int_0^\infty |A_2(f)|^2 df \Big/ \int_0^\infty |A_1(f)| \cdot |A_3(f)| df \quad (4)$$

and $|A_i(f)|$ are the Fourier transform magnitudes of $a_i(t)$, which are modeled as plane waves. In practice, the value of x is calculated in a frequency range where the magnitudes are above a predetermined level. For the examples illustrated in this work, the emitter has a central frequency of 5 MHz, and the frequency range used on equation 4 is between 1 and 9 MHz.

The use of the large-aperture receiver reduces the errors in the signals amplitude measurement, contributing to a more accurate measurement of the reflection coefficient.

2.1.2. Experimental results

Temperature plays an important role on density measurement, because it modifies cell dimensions and propagation velocities. It is assumed that the cell will operate in ambient temperature conditions, from 15 to 40°C. Buffer rod 1 is made of borosilicate glass, which has small thermal expansion coefficient [4.5 μm/(m.°C)], and its dimensions can be considered approximately constant in the temperature range considered.

Sample chamber length, L_2, is measured as a function of temperature, using distilled-water sample, whose propagation velocity is tabulated [13]. The cell is immersed in a thermostatic water bath and after the cell temperature stabilizes, the sample chamber length is measured. This process was repeated for a range of temperatures, changing in steps, resulting in a curve of L_2 as a function of temperature. For the cell used in this work, the slope of this curve is Δ_L=0.2 m/°C [14]. Then, the sample chamber length at a temperature T can be obtained from the measurement of $L_{2(cal)}$ at a single calibration temperature T_{cal}, from:

$$L_2(T) = L_{2(cal)} + \Delta_L (T - T_{cal}) \quad (5)$$

In Fig. 3 it is illustrated the measurement of the propagation velocity in distilled water as a function of temperature, with (o) and without (x) the sample chamber length correction, and when temperature is varied in steps of 5°C. In this case, calibration was made at the temperature of 20°C. The percent errors are measured by comparing the experimental and tabulated values, and the smaller errors when the compensation is used can be clearly seen. In this case, the transient errors in the measurement of propagation velocity, obtained while the temperature is not stabilized, are smaller than 0.07%, and for stabilized temperature, are smaller than 0.02%.

Table 1. Density of liquids obtained with the ultrasonic measurement cell and a pycnometer. Density values in kg/m^3.

Liquid	Density Measurement cell	Density Pycnometer	Relative difference [%]
Gasoline	747.6 ± 0.6	746.2	+0.19
Diesel	844.5 ± 0.6	842.7	+0.20
Alcohol	807.6 ± 0.5	808.6	-0.12
Corn oil	916.2 ± 0.8	917.0	-0.09
Glycerin	1260.2 ± 1.0	1260.3	-0.01

The density of several liquids was measured, under stabilized temperature conditions, at 20 ± 0.5°C [9]. The density values obtained with the cell were compared to the ones obtained with a pycnometer, and the results are shown in Tab. 1 [9]. Considering the pycnometer as a reference method, the errors in density measurement are smaller than 0.2% with conditions of stabilized temperature, and the standard deviations are smaller than 0.1% of

the mean value. Experiments with temperature gradients and distilled water showed that, while the temperature is varying (for example after a temperature step), the density is measured with an accuracy of 0.4%.

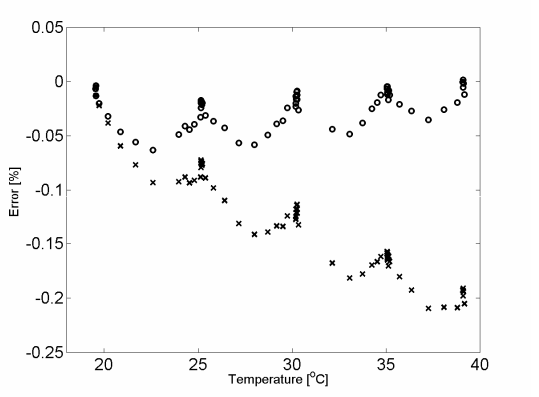


Fig.3 Percent errors in the propagation velocity in distilled water with (o) and without (x) correction in the sample chamber length.

2.2. Elastic constants

An anisotropic material is described by 21 independent elastic constants. The number of elastic constants is reduced to nine when the material has orthotropic symmetry. This number may be further reduced when there is more symmetry in the material. A unidirectional carbon-fiber/epoxy laminate can be considered as transversely isotropic and the number of independent elastic constants is reduced to five [10].

The determination of elastic constants from a set of bulk ultrasonic wave phase velocities in an arbitrary direction of a measured sample of composite material is based on the Christoffel's equation [15]. The use of large aperture PVDF receivers allows the measurement of plane wave in a wide range of frequencies, eliminating the need of diffraction correction. Besides, these thin-film piezopolymers have a relatively flat frequency response in a wideband of frequencies.

The diffraction effect is analyzed, in the range of 1 to 10 MHz, using aluminum, which is a low dispersion material [16]. The velocity dispersion is measured in a PMMA (polymetilmethacrylate) and a unidirectional CFRP samples [17].

2.2.1. Diffraction and dispersion on velocity measurement

The experiments have been made in a goniometer device immersed in distilled water. The water inside the goniometer was kept at 21.5 ± 0.05°C using a thermostatic bath. The goniometer device allows changing the emitter and the receiver transducers, as shown in Fig. 4. To analyze the diffraction effect, measurements of longitudinal velocity in a 9.5-mm thickness aluminum plate were conducted with 5 pairs (emitter and receiver) of 19-mm diameter NDT transducers (Panametrics model Videoscan) and frequencies of 1.0, 2.25, 3.5, 5.0 and 10.0 MHz. The receiver transducer is placed in the position of the PVDF receiver shown in Fig. 4.

The longitudinal velocity measurements in the aluminum plate were repeated using the same set of emitter transducers but using a large aperture PVDF receiver as shown in Fig 4.

The emitter is excited with one cycle sinusoidal wave (center frequency of transducer) using a function generator and a broadband power amplifier, and the electrical signals of the received waves are amplified (Panametrics 5072PR) and digitized by an oscilloscope (HP 500MHz bandwidth, 2Gs/s) connected to a computer via Ethernet. The echoes are stored and processed in the computer using MATLAB.

Fig.4 Photo of the goniometer

The large-aperture receiver is a 52-μm-thick PVDF membrane with gold electrodes. The PVDF membrane is bonded to a matched backing material, stiff enough to prevent low frequency bending vibration. The backing material has almost the same acoustic impedance of water around 20°C. The PVDF membrane is slightly stretched by using two concentric metallic rings. Each electrode is electrically connected to the corresponding terminal by contact rings. The external ring is grounded and the internal ring is connected to the signal.

Fig.5 Large-aperture PVDF receiver.

The effective diameter of the receiver was chosen in order to intercept the entire ultrasonic field produced by the transmitter. The PVDF receiver used in this work, shown in Fig. 5, has 80-mm active diameter.

Fig. 6 shows the measured longitudinal velocities in the aluminum plate [16]. The time delay between the two echoes, with and without sample, is measured using the Hilbert transform of the cross-correlation between them [9]. The results show that the diffraction effect may produce more than 1% error when using pair of transducers with 19-

mm diameter and 2-MHz frequency. This effect is eliminated with the large-aperture PVDF receiver as shown by the results of the dotted line and the result shown by the square mark.

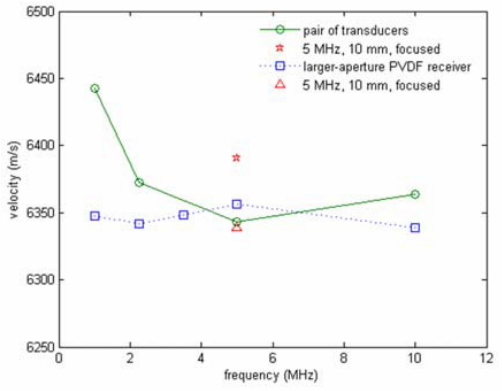


Fig.6 Measurements of longitudinal velocity in a 9.5-mm-thick aluminum plate.

Fig. 7 and 8 show the results of the longitudinal velocity measurement in an acrylic 4.5-mm-thick plate and a 2.115-mm-thick CFRP plate using a set of five NDT transducers of 1.0, 2.25, 3.5, 5.0, and 10.0MHz, 19-mm diameter, as emitters and the diffraction-free PVDF receiver [17].

The results show that the longitudinal velocity increases with the frequency, producing errors of about 1% in the acrylic and the CFRP plates when the frequency increases from 1 to 10MHz.

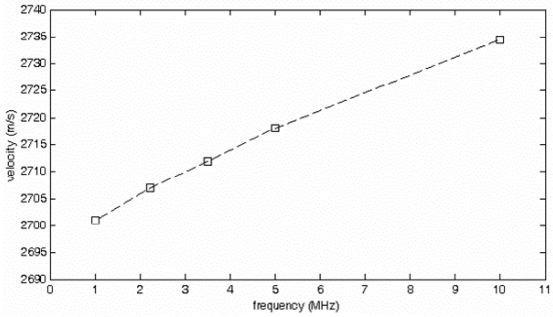


Fig.7 Measurements of longitudinal velocity for a 4.5-mm-thick plate.

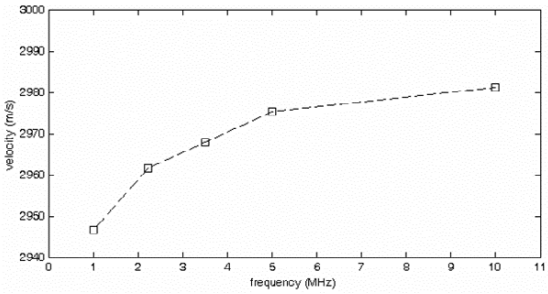


Fig.8 Measurements of longitudinal velocity for a 2.115-mm-thick CFRP plate.

2.2.2. Determination of elastic constants

Figure 9 shows the coordinate system attached to a unidirectional laminate. The fibers are placed parallel to axis x_3. The plane x_1-x_2 can be considered isotropic [10].

The phase velocity at a refraction angle θ_r, shown in Fig. 10, is obtained by measuring the time delay Δt of the ultrasonic wave traveling with and without the composite plate at a known temperature.

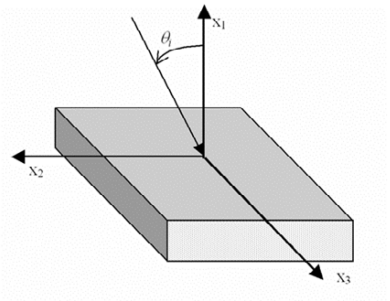


Fig.9. Coordinate system of a unidirectional laminate.

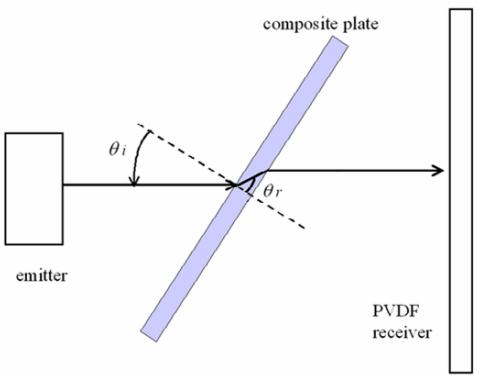


Fig.10 Schematic of the goniometer.

The acoustic velocity in water, v_w, is tabulated and can be obtained from the temperature. The phase velocity for a composite plate with thickness h is [11]:

$$v(\theta_r) = \left(1/v_w^{\,2} - 2\Delta t \cos\theta_i / h v_w + \Delta t^2 / h^2\right)^{-1/2} \tag{6}$$

where:

$$\theta_r = \sin^{-1}\left(v(\theta_r)\sin\theta_i / v_w\right) \tag{7}$$

and θ_i is the incidence angle.

The unknown elastic constants are found by minimizing an objective function F, which is the sum of the squares of the deviations between the experimental and calculated phase velocities, given by:

$$F = \sum_{i=1}^{N} (v_i^{\exp} - v_i^{calc})^2 \qquad (8)$$

where $v_i^{\exp}$ is obtained by (6), and N is the number of measured velocities. The minimization of F is implemented using *fminisearch* function of MATLAB.

2.2.3. Experimental results

The experimental results of the phase velocities were obtained from a 2.11-mm-thick unidirectional carbon-fiber/epoxy square plate (80 x 80mm) with density ρ=1585 kg/m^3 using a goniometer shown in Fig. 4 and Fig. 10.

The emitter is a focused transducer with nominal frequency of 5 MHz and diameter of 10mm, and the receiver is the diffraction-free PVDF transducer shown in Fig. 5. The velocities are measured in the plane x_1-x_2 and x_1-x_3, changing the incidence angle from 0 to 40°, spaced by 0.1°. The results are shown in Fig. 11.

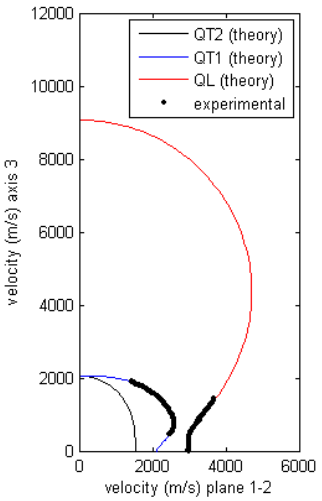


Fig.11 Phase velocities in the planes x_1-x_2 and x_1-x_3.

Table 2 shows the calculated results of the elastic constants obtained by minimizing function *F* from (8).

Table 3 shows the engineering constants (Young and shear modulus and Poisson's ratio) calculated from the elastic constants of table 2 and those obtained by a tensile test of samples of the same CFRP plate.

Table 2. Elastic Constants

Elastic constant	(GPa)
C_{11}	14.1
C_{12}	6.8
C_{13}	6.2
C_{33}	135
C_{44}	6.8

Table 3. Engineering Constants.

Constant	Calculated	Tensile test
E_1 (GPa)	10.7	10
E_3 (GPa)	132	140
G_{13} (GPa)	6.8	7
ν_{31}	0.30	0.31

3. Conclusions

Large-aperture PVDF receivers have broadband frequency response and are very useful for measurements of pressure amplitude and velocity without diffraction effects. Gathering signal processing techniques and carefully designed large-aperture receivers, resulted in a liquid density measurement with an accuracy of 0.2% under stabilized temperature conditions. Experimental results of diffraction effects on velocity measurement, in a low attenuation material, show that this error can be more than 1% when using a pair of 19-mm diameter transducers under 2 MHz. On the other hand, the measurements with the large-aperture PVDF receiver show the elimination of the diffraction effect, even when using a 5-MHz 10-mm diameter focused transducer. The dispersion effect is shown for the measurement of longitudinal velocity in an acrylic plate (medium attenuation) with the diffraction-free receiver. The fiber/plastic reinforced composites generally have high dispersive effect. The results of the elastic constant measurement with the large-aperture PVDF receiver show good agreement with the tensile test.

Acknowledgements

This research is based upon work supported by CNPq and PETROBRAS/ANP.

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